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Sorption of organic gases in residential rooms

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Abstract

Experiments were conducted to characterize organic gas sorption in residential rooms studied "as-is" with furnishings and material surfaces unaltered and in a furnished chamber designed to simulate a residential room. Results are presented for 10 rooms (five bedrooms, two bathrooms, a home office, and two multi-function spaces) and the chamber. Exposed materials were characterized and areas quantified. A mixture of volatile organic compounds (VOCs) was rapidly volatilized within each room as it was closed and sealed for a 5-h Adsorb phase; this was followed by 30-min Flush and 2-h closed-room Desorb phases. Included were alkane, aromatic, and oxygenated VOCs representing a range of ambient and indoor air pollutants. Three organophosphorus compounds served as surrogates for Sarin-like nerve agents. Measured gas-phase concentrations were fit to three variations of a mathematical model that considers sorption occurring at a surface sink and potentially a second, embedded sink. The 3-parameter sink-diffusion model provided acceptable fits for most compounds and the 4-parameter two-sink model provided acceptable fits for the others. Initial adsorption rates and sorptive partitioning increased with decreasing vapor pressure for the alkanes, aromatics and oxygenated VOCs. Best-fit sorption parameters obtained from experimental data from the chamber produced best-fit sorption parameters similar to those obtained from the residential rooms.

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1. Introduction

Indoor air pollution caused by indoor and outdoor sources of volatile organic compounds (VOCs) is impacted by a number of factors including the

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emission characteristics of the sources, building outdoor air exchange, interaction of the compounds with indoor surfaces and materials, and other loss mechanisms including reactive chemistry and air cleaning (e.g., Tichenor and Sparks, 1996). The importance of surface interactions cannot be overlooked. Adsorption of VOCs to surfaces and their diffusion into porous materials can significantly reduce peak concentrations and alter their temporal profiles in buildings. Such impacts are most likely to

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be apparent following an episodic release of a VOC indoors. Examples include the accidental or intentional release of a toxic chemical (Singer et al., 2005a), cleaning activities involving solvent-containing products (Singer et al., 2006), and other intermittent source events such as smoking (Singer et al., 2003). Sorption processes also can alter the concentration profile of diurnally-varying ambient VOC sources such as motor vehicle exhaust emissions that enter buildings through ventilation and infiltration. Peak concentration reduction may be significant even for compounds generally considered to be entirely volatile (e.g. ethylbenzene, limonene, pyridine) when the rate of adsorption to surfaces competes with loss due to air exchange (Singer et al., 2004). Subsequent re-emission of VOCs from surfaces and materials temporally redistributes occupant exposure potentials to later time periods. The health significance of a reduction in peak concentration may be exponential rather than linear, i.e. for compounds having a toxic load exponent greater than one (Singer et al., 2005a).

Recognition of the potential impact of sorption and other interactions with materials (e.g. diffusion) on indoor VOC exposures has stimulated interest in the development of tools and empirical data to quantify these processes. Indoor air quality models have been adapted to predict indoor VOCs exposures based on different combinations of sources and sinks (Tichenor et al., 1991; Tichenor and Sparks, 1996). Sink/diffusion models have been developed to characterize the interactions of VOCs with porous indoor surfaces (Jorgensen et al., 2000; Zhao et al., 2002). Sorption of selected VOCs on common indoor materials (e.g., carpet, gypsum wallboard, vinyl flooring) has been studied in small-scale chamber (<1 m³) experiments (Jorgensen et al., 1991; Tichenor et al., 1991; Won et al., 2001a; Zhang et al., 2002). The materials used in these experiments were aged by exposure to clean air and studied individually. Sorption models were fit to measured data to infer rate parameters for specific compound-material interactions. These interactions can be combined mathematically to simulate sorption in an entire room or house, as reported in a few studies, e.g. (Singer et al., 2004; Tichenor et al., 1991; Won et al., 2001b).

Our group has demonstrated another approach in which adsorption and re-emission of VOC mixtures have been studied directly in a room-sized, 49.5 m³ chamber finished with common materials and containing aged furnishings to simulate a residential

room (Singer et al., 2002, 2004, 2005a). This approach yields average or 'bulk' sorption parameters that may typify many residential rooms. Results indicate that adsorption and re-emission to an entire room can be characterized using semi-empirical models assuming either a surface sink or an embedded, e.g., diffusive sink. The dynamic behavior of low sorbing compounds (e.g., 2-butanone, benzene, toluene) were adequately described with the one-sink model, whereas the dynamics of highly sorbing compounds (e.g., nicotine, o-cresol, 1-methylnaphthalene) were more closely reproduced with the two-sink model (Singer et al., 2004).

Since rooms in occupied residences differ substantially in size and character and contain many varied materials whose exposed surfaces are difficult to characterize, it was deemed important to conduct corollary experiments in actual residences. This study was undertaken to determine sorption rate parameters for a mixture of representative VOCs in a convenience sample of rooms of different sizes and functions and to compare these parameters to those developed from experiments in the large-scale chamber. To our knowledge this is the first study to report measurements of sorption in a sample of actual residences.

2. Research methods

2.1. Overview

Sorption was studied by conducting controlled experiments in actual residential rooms and an experimental chamber at LBNL that was constructed, finished and furnished to simulate a residential room. Each experiment included rapid volatilization of VOCs and manipulation of air exchange to achieve periods of net adsorption and net desorption. Gas-phase concentrations were measured at appropriate intervals to capture their dynamics throughout these periods. Example data are shown in Fig. 1. The measured concentrations were fitted to semi-empirical sorption models to determine parameter values.

2.2. Study rooms

The research team and colleagues volunteered a sample of residences and rooms. Work was conducted when the residences were unoccupied for at least 10 h. Candidate rooms were those that

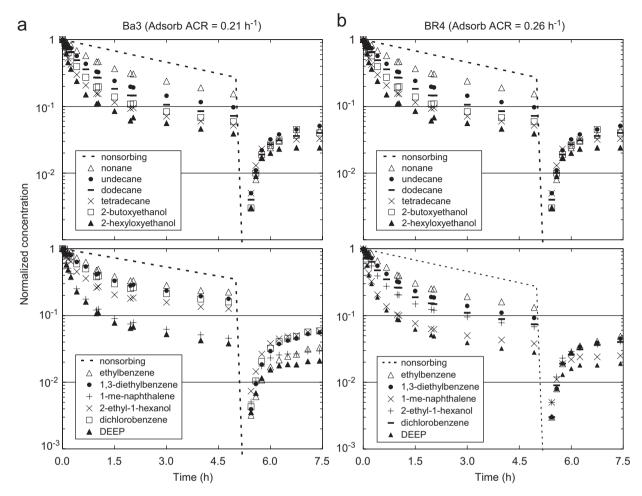


Fig. 1. Example data from Experiments Ba3 and BR4 for selected compounds. Rooms were closed and sealed to reduce air exchange during first 5 h (Adsorb phase). Rooms were rapidly ventilated between 5 and 5.5 h, then closed again from 5.5 to 7.5 h (Desorb phase).

reasonably could be sealed and isolated from adjoining spaces. Room selection aimed to achieve variation in assumed key characteristics including total volume, total material surface area, surface-to-volume ratio, and relative amounts of various types of materials. In total, experiments were conducted in 11 rooms (Table 1).

For each room and the test chamber, the volume and the apparent areas of material surfaces exposed to freely moving room air were characterized (Hodgson et al., 2004). Materials were grouped into four categories: (1) fleecy/padded (e.g., carpet, upholstery, bedding, other fabrics); (2) painted wallboard/plaster; (3) hard/porous (e.g., wood, plastic, laminate, paper); and (4) hard/nonporous (e.g., glass, metal, tile, porcelain). Room volumes presented in Table 1 and used to calculate surface to volume ratios exclude solid objects and, thus, are

estimates of the air mixing volume. The chamber was furnished with hard and plush materials as described previously (Singer et al., 2005a); the chamber air mixing volume was estimated to be 47.3 m³.

2.3. Experimental protocol

The experimental protocol was a modified version of one described previously (Singer et al., 2004, 2005a). Each experiment examined VOC adsorption and re-emission in a single room or contiguous space. Rooms were studied as found without cleaning or organization except that major air leakage openings such as heating supply ducts and large gaps around doors and windows were covered. This treatment reduced air exchange and thus improved the resolution of sorption processes.

Table 1 Summary of experiments in residential rooms and furnished chamber

Exp. ID	Room type	Vol. (m³)	ACR (h ⁻¹) Ads/Flush/Des	Adsorb T (°C)	Adsorb RH (%)	Desorb <i>T</i> (°C)	Desorb RH (%)
Ba1	Bathroom	6.7	$0.24/85^{a}/1.18^{b}$	n.m. ^c	n.m.°	n.m. ^c	n.m. ^c
Ba2	Bathroom	11	$0.36/6.2^{\rm d}/0.44$	23.3-27.3	46-54	24.8-27.7	44-49
Ba3	Bathroom	12	0.21/33 ^a /n.m. ^e	21.6-24.2	42-59	23.8-25.1	38-42
BR1	Bedroom	19	$0.16/16^{a}/0.20$	20.2-25.4	45-57	25.4-27.5	41–44
BR2	Bedroom	22	$0.08/23^{a}/0.08$	18.5-20.4	65–70	19.9-20.6	61–68
BR3	Bedroom	27	$0.11/46^{a}/0.17$	20.1-22.1	46-55	21.1-23.7	45-50
BR4	Bedroom	31	$0.26/32^{a}/0.26$	20.1-21.7	53-60	21.7-23.0	49-53
BR5	Bedroom	49	$0.15/27^{a}/0.17$	21.0-25.1	53-60	23.5-25.9	52-55
Off	Home office	29	$0.12/30^{a}/0.13$	21.9-22.5	56-59	22.1-22.8	55-56
FRK	Family room + kitchen	58	0.16/10/0.08	20.0–21.9	52–56	21.3–22.9	51–53
Eff	Efficiency apartment	65	0.19/10/0.18	16.0-26.3 ^f	$55 - 80^{\mathrm{f}}$	25.4–26.8	44–46
EC	Experimental chamber	47 ^g	0.16/5.9/0.17	20.1–20.9	59–63	20.8-20.8	58–59

^aReported high air change rates $(>15\,h^{-1})$ have large uncertainties. These should not affect fitting for determination of sorption parameters because removal of compound mass is limited by rate of desorption from material surfaces into air (by λ_d , k_{dif} or k_2).

Temperature and relative humidity were unregulated.

A VOC mixture was rapidly volatilized into the space establishing time zero (t = 0). This was followed by a net movement of VOC mass from the gas-phase to room materials (Adsorb phase, Fig. 1). The Adsorb phase lasted for 5 h (6 h in room FRK) allowing gas-phase concentrations to reach, or approach, a plateau indicative of quasi steadystate partitioning. The room was then ventilated for 30 min, typically by exhausting air through an open window using a large box fan (Flush phase), while another fan oscillated on high speed to promote mixing within the room. This rapidly removed compound mass remaining in the gas phase at the end of the Adsorb phase. During the Flush phase, the room was occupied by 1-2 researchers, who removed the Adsorb phase sorbent samplers, inserted sorbent tubes for the Desorb phase, and collected a Flush phase sample, all with the door closed (to minimize transport of compound mass into other parts of the residence). At $t = 5.5 \,\mathrm{h}$, the room was resealed and the increase in gas-phase VOC concentrations due to desorption of previously adsorbed mass was measured (Desorb phase). The Desorb phase lasted for 2 h, after which the room was ventilated at a high rate to facilitate removal of residual compound mass prior to reoccupancy.

2.4. Study compounds

The VOCs selected for study were: (1) four normal alkane hydrocarbons (nonane, undecane, dodecane, tetradecane); (2) four aromatic hydrocarbons (ethylbenzene, 1,2-dichlorobenzene, 1,3-diethylbenzene, 1-methylnaphthalene); (3) four oxygenated compounds including three with multiple functional groups (2-butoxyethanol, 2-hexyloxyethanol, 2-ethyl-1-hexanol, methyl salicylate); and (4) three organophosphorus (OP) compounds (dimethyl methylphosphonate (DMMP), diethyl ethylphosphonate (DEEP), triethylphosphate (TEP)). The first three groups of chemicals were intended

^bRoom has louvered window that was covered and sealed during Adsorb phase, but not sealed during Desorb phase.

 $^{^{}c}$ n.m. = not measured

 $^{^{\}rm d}6\,h^{-1}$ was fitted SF₆ decay for entire Flush phase. Decay rate of $12\,h^{-1}$ measured over first 7 min produced closer match to organic gas concentrations measured at end of Flush phase.

^eACR was not measured during Desorb phase. When fitting data to models, ACR was assumed to be the same as measured during Adsorb phase.

Most of variation occurred over first 50 min during which temperature rose from 16 to 25 °C and RH dropped from 80% to 49%.

^gVolume of empty chamber is 49.5 m³. Volume occupied by furnishings, primarily four upholstered chairs and a desk was 2.2 m³. The net air mixing volume was 47.3 m³.

to represent a variety of commonly encountered indoor air pollutants. Within each chemical group, the compounds spanned ranges of molecular mass and volatility (Table S1, Supplemental material). The three OP compounds have been used as surrogates to estimate absorption of Sarin-like nerve agents, and methyl salicylate has been used as a surrogate for mustard gas (Singer et al., 2005a).

Aliquots of neat compounds were combined in a small conical vial in sufficient quantities to achieve initial target concentrations of approximately $500-1500 \,\mu\mathrm{g}\,\mathrm{m}^{-3}$ in room air. The vial was sealed and transported cold to the study site. At the beginning of an experiment, the vial was opened in the room and the entire contents were drawn quickly into a syringe. The mixture was immediately injected into a glass dish preheated on a hot plate to ~180–240 °C. A household fan (high setting) circulated air across the dish towards the middle of the room. After 1 min, the hot plate was turned off, and the fan was set to oscillate (low setting) for the entire Adsorb phase. The researcher exited with the vial, syringe and glass dish. Then the gap under the door was covered and the room remained closed throughout the Adsorb phase.

2.5. Air sampling

Air samples were collected primarily using a timeprogrammable multi-port sampler onto sorbent tubes containing Tenax®-TA. A 15-cm section of 1.6-mm ID Teflon tubing was attached to the inlet of each sorbent tube to minimize passive sampling via diffusion during the standby time. A sampling rate of $\sim 100 \,\mathrm{ml\,min^{-1}}$ was maintained by electronic mass flow control and the device output voltage indicating flow rate was logged. Sample durations and intervals were set to maximize resolution of temporal concentration changes. Routinely, 15 samples were collected during the Adsorb phase at 3-4, 6-7, 9-10, 14-15, 24-25.5, 39-41, 57-60, 60-63, 88–92, 115–120, 120–125, 177–183, 236–243, 284–291, and 292–300 min, yielding sample volumes of ~0.1-0.8 L (timing of later samples differed slightly for Exp. FRK). Operational blanks were obtained by loading a sorbent tube into one port of the sampler that was not set to draw a sample; these blanks were collected during the Adsorb phase of all but one experiment (FRK), and in the Desorb phase of three experiments (Ba2, BR5, Off). Eight Desorb phase samples of 10 min duration (~1-L volume) each were initiated at 0, 10, 25, 40, 55, 70, 90 and

110 min after the room was resealed following the Flush phase.

Additional sorbent tube samples were collected in the room before the experiment and during the final 10 min of the Flush phase at ~100 ml min⁻¹ using a peristaltic pump. Other samples were collected in the same fashion in another room of the residence and/or outdoors to screen for external sources of study compounds. During the experiment in the chamber, samples were collected using the programmable automated sampler placed in the chamber and manually by inserting sample tubes through ports in the chamber wall. Samples were analyzed for the target compounds by thermal desorption gas chromatography/mass spectrometry as described previously (Singer et al., 2004, 2005a).

2.6. Other measurements

Air change rates (ACRs) were determined by the measured decay of SF₆ tracer gas. SF₆ was injected into a room just prior to product volatilization, at the beginning of the Flush phase, and just prior to the Desorb phase. Time-resolved SF₆ concentrations were measured using a photoacoustic infrared analyzer (Model 1302, Bruel & Kjaer). Temperature and relative humidity (RH) were monitored using data-logging sensors (HOBO H8 Pro, Onset Computer Corp.) set to record values every 5 min. Data from two or three T/RH sensors were averaged to provide a temporal profile in each experiment.

2.7. Modeling

Measured gas-phase concentrations were fitted to a mass balance sorption model (Eqs. (1)–(3)) that tracks mass in room air (C), in the exposed surface sink (M), and potentially in an embedded sink (E) in contact with the surface but not directly with room air. Concentrations C, M, and E are parameterized as the mass in each compartment divided by the room air volume (μ g m⁻³). Other parameters are the ACR λ (h⁻¹) and coefficients λ _a, λ _d, k₁, and k₂ (h⁻¹) describing rates of mass transfer among compartments (Singer et al., 2005a):

$$\frac{\mathrm{d}C}{\mathrm{d}t} = -(\lambda + \lambda_{\mathrm{a}})C + \lambda_{\mathrm{d}}M,\tag{1}$$

$$\frac{\mathrm{d}M}{\mathrm{d}t} = \lambda_{\mathrm{a}}C - (\lambda_{\mathrm{d}} + k_{1})M + k_{2}E,\tag{2}$$

$$\frac{\mathrm{d}E}{\mathrm{d}t} = -k_2 E + k_1 M. \tag{3}$$

Eqs. (1)–(3) are analogs to those used to describe sorption and diffusion of VOCs interacting with single materials in small chambers. Our approach groups all furnishings and room surfaces into one or two conceptual sinks. The parameterization of sorption coefficients as rates (h⁻¹) allows direct comparison between sorption and air change time scales. In other formulations, the adsorption coefficient has velocity units (i.e., m h⁻¹) and sorbed mass concentration is normalized to apparent surface area (i.e., µg m⁻²). These formulations differ from our λ_a and M by the apparent surface area to air volume ratio. Relating the mass in each sink to room air volume allows for straightforward evaluation of mass partitioning. Eqs. (1)–(3) can represent three conceptual sorption models. The sink model $(k_1 = k_2 = 0)$ assumes sorption at the surface only. The sink-diffusion model adds a simplified diffusion mechanism at rate $k_1 = k_2 = k_{\text{dif}}$ between surface and embedded sinks (Jorgensen et al., 2000). The two-sink model allows unequal rates of mass movement between surface and embedded sinks $(k_1 \neq k_2)$. Parameter values are obtained by fitting the models to measured gas-phase concentrations. Partitioning coefficients (K_{part}) defined as the ratio of sorbed- to gas-phase mass at equilibrium, were calculated from rate coefficients as described previously (Singer et al., 2005a).

A goodness-of-fit (GF) metric was calculated as the root mean square of normalized residuals divided by the square root of N measured points (Eq. (4)). Normalized residuals were calculated from measured (y_i) and model-predicted (y_i^*) values:

GF =
$$\sqrt{\frac{\sum ((y_i - y_i^*)/y_i)^2}{N}}$$
. (4)

2.8. Data analysis

Best-fit coefficient values for the models shown in Eqs. (1)–(3) were determined by minimizing the GFs using (1) all measured data and (2) Adsorb phase data only. In Exp. FRK, Desorb phase organic gas data were unavailable due to sampler malfunction; thus the models were fit to Adsorb data only. Concentrations at t=0 were calculated by backward extrapolation from the first 3–4 measurements of the Adsorb phase. Model fits were reviewed visually to screen for cases when the initial fit settled

on a localized minimum GF or the fit exhibited a systematic but compensating bias (e.g. consistent under-prediction on measured concentrations early and over-prediction later in Adsorb phase. Fits based on Adsorb phase only and all data were compared as another check of robustness. The two approaches generally yielded similar and consistent parameter values as reported previously for the OP compounds (Singer et al., 2005a). The parameter value results presented in this paper are mostly from the all-data fits. Exceptions are all compounds in Experiment FRK and nonane in Experiments Bal and BR5. When fitting to all data, a model was judged to be acceptable for GF ≤ 0.15 (Singer et al., 2005a), but other target values could be used. For undetermined reasons, acceptable and consistent model fits between Adsorb only and all data were not obtained for Experiment Ba2. Results from this experiment are not included in summary tables; however, the observed initial Adsorb phase decay rates are similar to those observed across other experiments.

Concentrations measured in the room prior to the beginning of each experiment were <10% of the final concentrations measured at the end of the Adsorb phase except for the following cases: nonane (Ba1, 18%; BR3, 16%); tetradecane (Ba1, 19%; FRK, 21%); 2-butoxyethanol (Ba1, 19%; BR2, 17%; BR3, 11%; BR4, 14%; Off, 10%). These compounds are ubiquitous indoor air pollutants and their presence in air at these levels is not surprising. No adjustment was made for concentrations measured prior to the start of experiments. Also, compound mass already sorbed to surfaces in the study rooms was ignored in the analysis.

VOC masses collected on each operational blank were compared to masses collected during the corresponding experimental periods. Masses collected on operational blanks were <5% of the lowest observed Adsorb phase sample masses for all compounds in Experiments Ba1, Ba3, BR1, BR2, BR3, BR4, FRK, and Eff. The final three experiments conducted (in chronological order) were Off, Ba2, and BR5. In these experiments, VOC blank masses were up to 15% of the lowest collected Adsorb phase sample masses. Diffusive sampling (during the sampler standby time) cannot account for these results. We hypothesize, but did not confirm, that this was caused by a small leak in the 16-port valve of the sampler possibly due to misalignment of the head. Adsorb phase data for Experiments Off, Ba2, and BR5 were adjusted by subtracting the operational blank masses from the collected masses.

3. Results and discussion

3.1. Study room characteristics

The characteristics of the study rooms and the chamber are summarized in Table 1 and Fig. 2. The rooms ranged in size from 7 to 65 m³ and exhibited substantial variability in the relative quantities of materials present. Overall material surface to volume (S/V) ratios ranged from 2.9 to $4.6 \,\mathrm{m}^{-1}$. In general, the smaller rooms had higher S/V ratios. Bathrooms were the smallest rooms studied and exhibited the highest overall S/V ratios, with 25–35% of the area in these rooms attributed to nonporous materials such as fixtures and tile. The home office (Off) was similar to the bathrooms in the overall S/V ratio and the relative paucity of plush materials. The largest two bedrooms had similar material distributions to the multipurpose rooms (FRK and Eff). Across the bedrooms and multipurpose rooms, S/V ratios for the most prominent material categories varied by factors of 1.5 for wallboard/plaster, 2.1 for other hard

surfaces, and 2.3 for plush materials. The furnished chamber had the lowest overall S/V ratio but had relative quantities of plush materials and wallboard similar to residential bedrooms and mixed use rooms. While the summary data provide general descriptions of the materials present in the rooms, it is important to note that the sorptive properties of the specific materials within each category—especially the plush materials group—may vary greatly. Also, the actual surface area of each material may be much larger than the apparent or measured area. This enhancement is greatest for plush materials.

3.2. Experimental conditions

Conditions in the field experiments varied among and sometimes during experiments. One important parameter, which was partially controlled and measured throughout each experiment, was the air change rate (ACR). Adsorb phase ACRs were in the range of 0.08–0.36 h⁻¹. These values were sufficiently low to allow for accurate estimation of the initial adsorption rate, a key model parameter. Very high ACRs were achieved during the Flush phase providing successful removal of compound mass remaining in the gas-phase at the end of the Adsorb

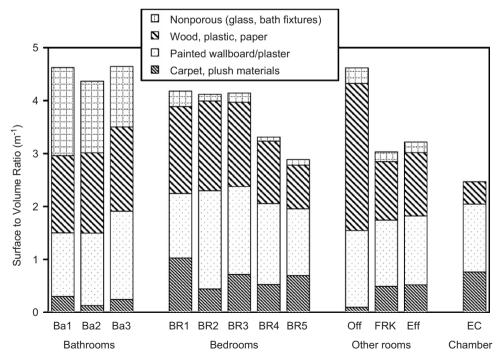


Fig. 2. Estimated surface to volume ratios in study rooms and chamber, accounting for predominant material categories present in study spaces.

period. In most cases, the ACR during the Desorb phase was similar to that measured during the Adsorb phase.

Temperature and RH data were recorded in 10 of the 11 residential experiments (Table 1). In seven of ten experiments, Adsorb phase temperature varied by $<3\,^{\circ}$ C, and Adsorb phase RH varied by <10%. In Experiment Eff, a significant increase in temperature and a concurrent decrease in RH occurred over the first 50 min. In Experiments Ba2 and BR1 most of the change in these parameters occurred within the first 90 min. Since higher temperatures drive sorptive partitioning to the gas-phase, a temperature increase during the early part of the Adsorb phase would be expected to suppress adsorption. To the extent that this occurred, values of λ_a from these experiments may be biased on the low side.

3.3. Measured organic gas concentrations

Fig. 1 presents example results for 10 compounds in two rooms (Ba3 and BR4) with similar Adsorb phase ACRs (0.21 and 0.26 h⁻¹) and illustrates several trends for the entire dataset. The calculated concentration pattern of a theoretical, nonsorbing compound is also shown for reference. The Adsorb phase includes an initial period in which the combined concentration decay rate is proportional to the sum of the ACR and the initial adsorption rate (λ_a) , followed by a period in which concentrations approach a plateau indicative of equilibrium partitioning. The order of the compounds is similar between the two experiments even though the decay rates differed somewhat. The most substantially sorbing compounds—including tetradecane, 2-hexyloxyethanol, 1-methylnapthalene, and DEEP (shown); and DMMP, TEP and methyl salicylate (not shown)—behaved similarly to one another within the Adsorb phase of each experiment. The observed initial decay of these compounds is thought to represent the transport-limited deposition rate for the extant conditions in each experiment.

Gas-phase concentrations of all compounds decreased to very low levels during the Flush phase and increased rapidly at the beginning of the Desorb phase. Concentrations of all species increased on a similar time scale during the Desorb phase suggesting that process may also be limited by transport rather than compound-material (i.e. sorbent–sorbate) interactions.

3.4. Best-fit sorption parameter values

Best-fit sorption parameter values are summarized by chemical class in Tables 2-5, and representative model fits are shown for four compounds in Fig. 3. In each table, results are presented for the 10 residential rooms and for the furnished chamber. Fitted parameters for the sink and sink-diffusion models are shown for all compounds. Two-sink model parameters are additionally shown for oxygenated VOCs and OP compounds (Tables 4 and 5). These results are discussed in relation to the questions that motivated the research: (1) can the lumped-material (whole-room) models in Eqs. (1)–(3), represent sorption dynamics observed in residential rooms; (2) how do the parameters determined from residential room experiments compare to those determined for the furnished chamber; and (3) what are the ranges and central tendency values for the parameters in residential rooms?

(1) Overall, the results in Tables 2–5 demonstrate that the whole-room models are applicable to residential rooms. The sink diffusion model reasonably fitted (GF \leq 0.17) the residential room data for all alkanes, aromatics, and oxygenated VOCs; and approximated observed dynamics for OP compounds with GFs in the range 0.19-0.25. Improved fits were obtained using the two-sink model for substantially sorbing compounds, as shown for oxygenated VOCs (Table 4) and OP compounds (Table 5). These results are generally consistent with model applicability determined in chamber experiments (Singer et al., 2004), with one notable exception. Previously, the sink-diffusion model was deemed sufficient for low- to moderately sorbing compounds and the two-sink model was required to provide acceptable fits for more rapidly and substantially sorbing compounds. Here "rapid" and "substantial" correspond roughly to initial adsorption rates (λ_a) above $1.5 \, h^{-1}$ and partitioning coefficients of approximately 10 or greater. Tetradecane, 1-methylnaphthalene, 2-hexyloxyethanol and methyl salicylate satisfy these conditions, yet they are reasonably well represented by the sinkdiffusion model in residential rooms. Some slightly higher GFs resulted when the sink-diffusion model was used for these compounds in the new chamber experiment (bottom half of Tables 2-5). The difference in sink-diffusion model applicability between these new data and previous chamber experiments may be related to the different protocols used, as examined later.

Table 2 Best-fit sorption parameters based on experiments in 10 residential rooms ($\mu \pm \sigma$) and furnished experimental chamber: results for *n*-alkane hydrocarbons

Compound	Model	$\lambda_a (h^{-1})$	$\lambda_{\rm d}~({\rm h}^{-1})$	$k_{\rm dif}~({\rm h}^{-1})$	GF^a	K_{part}
Experiments in 10 i	residential rooms					
Nonane	Sink	0.40 + 0.14	0.28 + 0.23		0.17	1.9 + 0.9
	Sink-dif	0.48 + 0.15	0.50 + 0.29	0.21 + 0.02	0.16	2.3 + 1.0
Undecane	Sink	0.82 + 0.19	0.19 + 0.04		0.15	4.4 + 1.3
	Sink-dif	$\frac{-}{1.04+0.23}$	0.45 + 0.15	0.24 + 0.08	0.11	5.0 + 1.6
Dodecane	Sink	1.12 ± 0.30	0.16 ± 0.03	_	0.18	7.4 ± 2.4
	Sink-dif	$\frac{-}{1.36+0.37}$	0.35 + 0.09	0.21 + 0.04	0.12	8.3 + 2.7
Tetradecane	Sink	2.6 ± 0.8	0.21 ± 0.06	_	0.19	13 ± 3
	Sink-dif	3.3 ± 1.0	0.49 ± 0.19	0.27 ± 0.08	0.12	14 ± 4
Experiment in chan	nber (plush+hard furn	ishings)				
Nonane	Sink	0.57	0.34		0.10	1.7
	Sink-dif	0.89	0.88	0.18	0.03	2.0
Undecane	Sink	0.86	0.14		0.12	6.0
	Sink-dif	1.03	0.29	0.18	0.08	7.1
Dodecane	Sink	1.06	0.11		0.18	9.6
	Sink-dif	1.22	0.22	0.18	0.13	11
Tetradecane	Sink	2.33	0.14		0.25	17
	Sink-dif	2.79	0.31	0.20	0.17	18

^aGoodness of fit metric (see Eq. (4)).

Table 3 Best-fit sorption parameters based on experiments in 10 residential rooms ($\mu \pm \sigma$) and furnished experimental chamber: results for aromatic compounds

Compound	Model	$\lambda_a \ (h^{-1})$	$\lambda_d (h^{-1})$	$k_{\rm dif}({ m h}^{-1})$	GF^a	K_{part}
Experiments in 10 rest	idential rooms					
Ethylbenzene	Sink	0.52 ± 0.17	0.18 + 0.12		0.16	3.4 ± 1.3
	Sink-dif	0.64 ± 0.21	0.36 ± 0.20	0.19 ± 0.02	0.13	$\frac{-}{4.1 \pm 1.4}$
Diethylbenzene	Sink	0.77 ± 0.19	0.14 + 0.03		0.17	5.6 ± 1.4
•	Sink-dif	0.92 ± 0.24	0.29 ± 0.07	0.19 ± 0.02	0.13	6.5 ± 1.7
1-me-naphthalene	Sink	3.0 + 0.9	0.16 + 0.03		0.24	19+5
T	Sink-dif	3.5 ± 1.0	0.34 ± 0.07	0.22 ± 0.02	0.15	21 ± 5
Dichlorobenzene	Sink	1.0 + 0.3	0.16 + 0.07		0.17	6.4 ± 2.1
	Sink-dif	1.2 ± 0.3	0.36 ± 0.20	0.24 ± 0.12	0.12	7.3 ± 2.5
Experiment in chambe	er (plush+hard furn	ishings)				
Ethylbenzene	Sink	0.7	0.27		0.12	2.7
•	Sink-dif	1.0	0.66	0.18	0.05	3.1
Diethylbenzene	Sink	1.0	0.15		0.15	6.7
•	Sink-dif	1.2	0.32	0.19	0.10	7.7
1-me-naphthalene	Sink	4.0	0.14		0.26	29
•	Sink-dif	4.5	0.29	0.21	0.16	31
Dichlorobenzene	Sink	1.4	0.16		0.19	8.4
	Sink-dif	1.7	0.35	0.19	0.12	9.5

^aGoodness of fit metric (see Eq. (4)).

Table 4
Best-fit sorption parameters based on experiments in 10 residential rooms ($\mu \pm \sigma$) and furnished experimental chamber: results for oxygenated volatile organic compounds

Compound	Model	$\lambda_a (h^{-1})$	$\lambda_d (h^{-1})$	$k_{\text{dif}}(\mathbf{h}^{-1}) \text{ or } k_1$ (\mathbf{h}^{-1})	k_2 (h ⁻¹)	GF^{a}	K_{part}
Experiments in 10 res	idential rooms						
2-Butoxyethanol	Sink	1.3 + 0.5	0.15 + 0.04			0.20	9.0 + 2.7
	Sink-dif	$\frac{-}{1.6+0.6}$	0.33 + 0.12	0.20 + 0.02		0.14	$\frac{-}{10+3}$
	Two-sink	2.4 ± 0.8	1.1 + 0.5	0.58 + 0.19	0.17 ± 0.08	0.08	11 ± 4
2-Ethyl-1-hexanol	Sink	$\frac{-}{1.4+0.4}$	0.18 + 0.03	_		0.19	8.0 ± 2.2
	Sink-dif	$\frac{-}{1.8+0.5}$	0.41 + 0.10	0.22 + 0.03		0.12	8.9 + 2.5
	Two-sink	$\frac{-}{2.5+0.8}$	$\frac{-}{1.1 + 0.4}$	0.56 + 0.22	0.20 ± 0.09	0.07	9.7 ± 3.6
2-Hexyloxyethanol	Sink	2.5 ± 0.9	0.16 + 0.03	_		0.25	$\frac{-}{16 \pm 4}$
	Sink-dif	2.9 + 1.0	0.33 + 0.07	0.21 + 0.02		0.17	17 + 4
	Two-sink	4.0 + 1.3	1.0 + 0.3	0.58 + 0.23	0.16 ± 0.06	0.10	19 ± 6
Methyl salicylate	Sink	2.7 ± 1.0	0.20 + 0.05			0.23	14+4
	Sink-dif	3.2 ± 1.1	0.44 ± 0.14	0.23 ± 0.03		0.15	15±5
	Two-sink	4.0 ± 1.4	0.93 ± 0.45	0.54 ± 0.25	0.22 ± 0.08	0.11	16 ± 5
Experiments in 10 res	ridential rooms						
2-Butoxyethanol	Sink	1.2	0.06			0.25	18
•	Sink-dif	1.3	0.12	0.17		0.21	21
	Two-sink	2.3	0.89	0.51	0.05	0.04	30
2-Ethyl-1-hexanol	Sink	1.7	0.12			0.24	13
•	Sink-dif	1.9	0.26	0.19		0.17	15
	Two-sink	3.0	0.87	0.39	0.09	0.05	18
2-Hexyloxyethanol	Sink	2.6	0.11			0.29	23
	Sink-dif	2.9	0.24	0.20		0.22	25
	Two-sink	4.1	0.45	0.36	0.14	0.19	33
Methyl salicylate	Sink	3.3	0.15			0.27	22
, ,,	Sink-dif	3.8	0.32	0.21		0.18	24
	Two-sink	5.0	0.83	0.45	0.13	0.06	26

^aGoodness of fit metric (see Eq. (4)).

(2) A comparison of the parameter values presented in upper and lower portions of Tables 2-5 shows that the adsorption rates (λ_a) and the equilibrium partitioning coefficients (K_{part}) derived from the chamber experiment are generally similar to the corresponding values determined for residential rooms. This is an important result as these parameters are the most robust, owing to the design of the experiments. The adsorption rate λ_a derives directly from the initial decay following compound volatilization and is therefore independent of later sorption dynamics. K_{part} is related to the steady concentration at the end of the Adsorb phase, especially for fast and substantially sorbing compounds. To facilitate the comparison of other parameters, Fig. 4 presents the best-fit values for all four parameters of the sink-diffusion model. The trend of parameter values across compounds is similar for both the rooms and the chamber, and the chamber values generally fall within the ranges of the room values. The two least-sorbing compounds, nonane and ethylbenzene, exhibited chamber-derived values for λ_a and λ_d that exceed the variances measured among residential rooms. Also, the λ_d and $k_{\rm dif}$ values determined in the chamber are consistently lower than those determined in residential rooms for oxygenated VOCs. Chamber-derived $K_{\rm part}$ values tend to be somewhat higher than room-derived values for these compounds. Still, these results indicate that parameter values obtained from experiments in the chamber are appropriate for estimating sorption in residential environments.

(3) Tables 2–5 indicate variability of sorption parameter values among residential rooms of roughly ± 25 –35% for λ_a and $K_{\rm part}$, ± 25 –50% for $\lambda_{\rm d}$, and ± 10 –20% for $k_{\rm dif}$. The uncertainty of individual parameters determined for a given experiment has been estimated to be of similar magnitude, i.e., roughly $\pm 30\%$ (Singer et al., 2004, 2005a).

Table 5
Best-fit sorption parameters based on experiments in 10 residential rooms ($\mu \pm \sigma$) and furnished chamber: results for organophosphorus (OP) nerve agent surrogates^a

Compound	Model	$\lambda_a (h^{-1})$	$\lambda_d (h^{-1})$	$k_{\rm dif} ({\rm h}^{-1}) { m or} k_1 $	$k_2 (h^{-1})$	GF^b	K_{part}
Experiments in I	10 residential rooms						
DMMP	Sink	2.9 ± 1.2	0.10 ± 0.03			0.31	31 ± 13
	Sink-dif	3.2 ± 1.3	0.20 ± 0.07	0.21 ± 0.03		0.25	34 ± 14
	2-sink	4.3 ± 1.6	0.74 ± 0.37	0.59 ± 0.25	0.11 ± 0.05	0.15	42 ± 18
DEEP	Sink	3.1 ± 1.1	0.13 ± 0.03			0.27	24 ± 9
	Sink-dif	3.5 ± 1.2	0.27 ± 0.07	0.21 ± 0.02		0.19	27 ± 9
	2-sink	4.7 ± 1.4	0.84 ± 0.34	0.58 ± 0.21	0.15 ± 0.04	0.09	29 ± 10
TEP	Sink	3.4 ± 1.3	0.14 ± 0.03			0.27	25 ± 10
	Sink-dif	3.8 ± 1.3	0.30 ± 0.07	0.22 ± 0.02		0.19	27 ± 11
	2-sink	5.0 ± 1.5	0.84 ± 0.23	0.58 ± 0.20	0.16 ± 0.05	0.11	29 ± 12
Experiments in I	10 residential rooms						
DMMP	Sink	2.5	0.08			0.31	30
	Sink-dif	2.7	0.17	0.19		0.25	33
	2-sink	4.3	0.86	0.54	0.08	0.07	39
DEEP	Sink	3.2	0.13			0.29	26
	Sink-dif	3.7	0.27	0.20		0.20	28
	2-sink	5.0	0.79	0.42	0.10	0.04	33
TEP	Sink	3.8	0.13			0.30	30
	Sink-dif	4.3	0.27	0.20		0.21	32
	2-sink	5.6	0.75	0.43	0.11	0.05	38

^aSelected as surrogates for G-series nerve agents including sarin and soman; DMMP: dimethylmethylphosphonate; DEEP: diethylethylphosphonate; TEP: triethylphosphonate.

A corollary question to those raised above is whether sorption dynamics vary with room type or as a function of the predominant materials contained within rooms. In Table S2 of the Supplemental Information, we provide parameter values for λ_a and K_{part} determined in the five bedrooms and the two bathrooms. While the numbers of rooms are small, there are no obvious differences between these two room types. Likewise, there were no significant correlations between sorption parameter values (e.g. λ_a or K_{part}) and room material characteristics (e.g. total S/V and S/V of plush materials).

The importance of individual sorption parameter values and their effect on indoor concentration profiles of chemical compounds has been discussed previously (Singer et al., 2004, 2005a). We again note the central importance of the initial adsorption rate (λ_a) in relation to the air change rate. When adsorption occurs at a rate that is competitive with or faster than air exchange, sorption will have a noticeable impact on time dependent concentrations. When adsorption is much faster than air exchange, it will serve as the primary removal

mechanism (from air) of a compound that is released in or infiltrates into a building. For example, the values of λ_a determined here for OPs in residential rooms are $\sim 4-5\,h^{-1}$ compared with typical infiltration or ventilation rates of $0.2-1.5\,h^{-1}$. This supports our previous assertion that adsorption is a key contributor to the efficacy of a shelter-in-place response to an outdoor nerve agent attack (Singer et al., 2005a). In addition, adsorption of OP compounds in rooms is so fast that this process likely would serve as an important removal mechanism for an indoor release (e.g. in an office building) under typical ventilation conditions.

The partitioning coefficient (K_{part}) determines the ultimate extent of sorption. Controlled experiments in the chamber have indicated that furnished rooms serve as large reservoirs for sorbing compounds (Singer et al., 2003). Thus, even for an ongoing or especially large release of contaminant, the vast majority of mass may partition to the sorbed phase if K_{part} is large. K_{part} values determined in this study demonstrate this for the OPs and some other compounds.

^bGoodness of fit metric (see Eq. (4)).

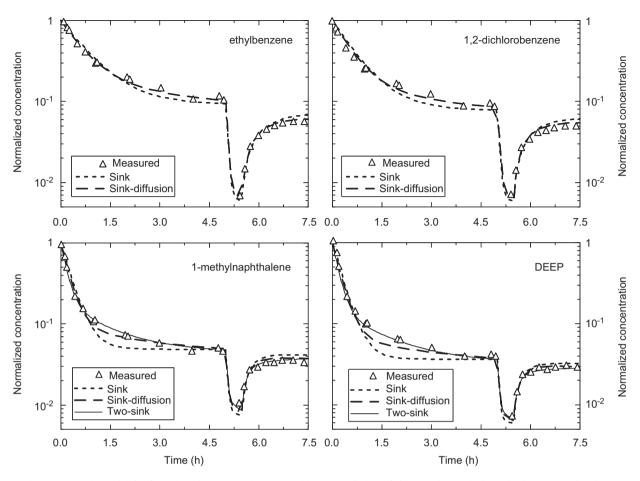


Fig. 3. Example model fits for selected compounds measured in BR1. Timing of the experimental phases is the same as in Fig. 1.

Another result that is consistent with previous findings is the consistency across models of the parameter controlling desorption (irrespective of the model fit). In the sink model, the rate of desorption is determined by λ_d . In the sink–diffusion model λ_d is often larger (i.e. faster) than k_{dif} ; this suggests that mass transfer back from the conceptual embedded sink to the surface is the process that limits the overall rate of desorption. In the two-sink model, the limiting desorption parameter typically is k_2 . The values obtained for λ_d in the sink-model, k_d in the sink-diffusion model and k_2 in the two-sink model are similar for each compound. Each of these parameters is also similar across compounds. And interestingly, the central tendency of these desorption-controlling rates is similar to the central tendency of the closed room air change rates, 0.2 h⁻¹. This result suggests that mass removal via desorption was limited by equilibrium partitioning. This hypothesis may be examined by varying the air

change rate during the Desorb phase in future experiments.

3.5. Extrapolation of results

Previous work has shown that for a given material (Won et al., 2001a), or furnished room where there is a mixture of materials (Singer et al., 2004), sorption parameter values can be related to compound vapor pressure to develop predictive curves by chemical class (see also Goss, 1997). Fig. 5 presents the relationship for λ_a values determined by applying the sink–diffusion model to the residential sorption data (Tables 2–5). We note that a similar plot included in an earlier conference paper (Singer et al., 2005b) was based on vapor pressure values that were incorrect (low) by 10^3 . Fig. 5 demonstrates that vapor pressure is a reasonably good predictor of the adsorption rate for VOCs spanning several classes, yet the rates for the OP nerve agent

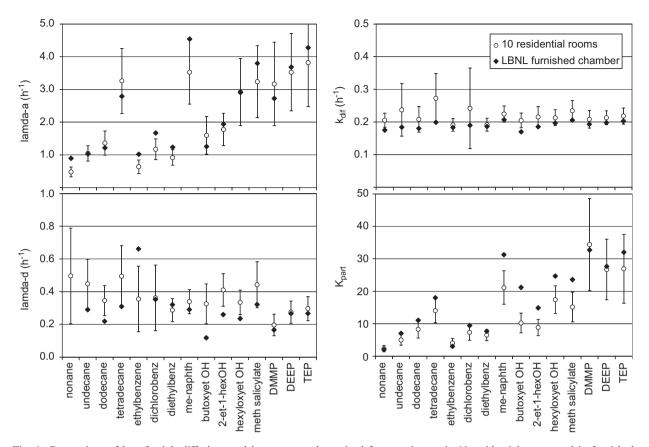


Fig. 4. Comparison of best-fit sink-diffusion model parameters determined for experiments in 10 residential rooms and in furnished chamber.

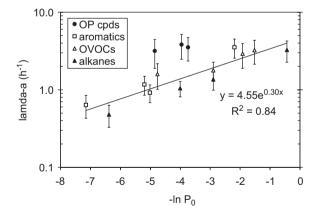


Fig. 5. Relationship between vapor pressure (P_0 (Pa)) and sink–diffusion model adsorption rates determined from room experiments. Fit shown is for all data except OP compounds.

surrogates are not well predicted by this relationship. An analogous plot for K_{part} provided in the Supplemental Information (Fig. S1) follows a similar trend.

3.6. Comparison of experimental protocols

The protocols employed in the current study differed in two respects from those used previously in the chamber (Singer et al., 2004, 2005a). (1) The new protocol used shorter Adsorb and Desorb phases of 5 and 2h, respectively, to facilitate completion of an experiment within the minimum 10h period over which the residence was unoccupied. These compare to Adsorb and Desorb phases of at least 22 and 8 h used previously. (2) ACRs during the room experiments were considerably higher than those previously achieved in the sealed chamber $(0.08-0.36\,h^{-1})$ versus $0.01-0.02\,h^{-1}$ for the Adsorb phase). In the new experiments, the chamber was intentionally ventilated at a rate of $\sim 0.16\,h^{-1}$ to simulate the room conditions.

In order to connect the new experiments with our previous work, we conducted several experiments in the chamber with the same mixture of compounds using both protocols. The experiments with the "long" protocol (extended Adsorb phase, lower ACRs) were previously described as Experiments

3-5 in Singer et al. (2005a). Surface-sink and sink-diffusion model results for alkanes, aromatics and oxygenated VOCs obtained from the full furnishing condition of Experiment 3 are presented in the Supplemental Information as Table S3. Twosink model results from the long and short protocol chamber experiments are shown for selected compounds in Table S4. OP results from Experiment 3 are presented in Table 7 of Singer et al. (2005a). Sorption parameter values obtained from the long protocol Experiment 3 followed similar trends by compound and model and many were similar in absolute terms to those obtained with the short protocol; this is especially true for λ_a . Systematic differences were observed for k_{dif} , k_2 and K_{part} . Values of $k_{\rm dif}$ obtained using the short protocol were consistently about $0.2 \, h^{-1}$ (Tables 2–5), whereas values from the long protocol experiment were about $0.05\,\mathrm{h^{-1}}$ (Table S3). Long protocol values of k_2 were $0.02-0.04 \, h^{-1}$ compared with short protocol values of $0.11-0.14 \, h^{-1}$ (Table S4). For substantially sorbing compounds (e.g., tetradecane, 1-methylnaphthalene, methyl salicylate), calculated K_{part} was higher with the long protocol compared to the short protocol owing directly to the lower λ_d and k_2 values. One possible explanation for the different k_{dif} and k_2 values obtained with short and long protocols is that the short Adsorb phase limits the time for sorptive partitioning and thus allows only a rough estimate of the slower processes. Another is that the rate of reemission may be limited by equilibrium partitioning between sorbed and gas-phase concentrations. By this explanation, the higher rates of desorption observed in the fast protocol are related to the higher ACRs in those experiments. This question may be investigated in future experiments.

The advantage of the short protocol is that it allowed study of sorption in actual residential rooms under low but realistic ventilation conditions. The flash volatilization protocol provided an emission and adsorption pattern directly relevant to episodic release events such as use of a VOC-containing cleaning product or the occurrence of an indoor or outdoor toxic chemical release. VOC dynamics in these scenarios are expected to closely follow those observed directly in experiments with the short protocol.

4. Conclusions

The goals of this work were to determine and compare sorption parameters for a variety of VOCs

in residences and to compare these results to those obtained in a furnished chamber simulating a residential room. Implicit in these goals was the need to validate that lumped parameter models are appropriate for use in modeling of sorption in residential rooms and that a newly developed protocol was appropriate for the determination of sorption parameters. The lumped parameter model(s) acceptably represented the observed sorption dynamics for all compounds studied. A two-parameter surface-sink model was sufficiently detailed to roughly track behavior of several lower-sorbing VOCs and a three-parameter sink-diffusion model matched the profiles for most of the more strongly sorbing compounds. A four-parameter two-sink model provided excellent fits even for the most strongly sorbing compounds. The initial adsorption rate λ_a is the most robust and accurate of the parameters obtained and together with the partitioning coefficient K_{part} provides the most valuable information for assessing the potential impact of sorption on acute exposures following a short-term release event.

Sorption varied moderately across the rooms studied without a clear trend by room type or amounts of materials present. Rather, compound vapor pressure was found to be the largest determinant of sorptive behavior. For all compounds studied, sorption occurred rapidly enough to compete with or exceed ventilation as the most important removal process following an indoor release.

The furnished chamber appears to be a representative model space for studying VOC sorption relevant to residential environments. Experimentation in the chamber has the obvious advantage of allowing control over important parameters such as ventilation rate, temperature, and RH, and allows for characterization of processes occurring over periods of tens of hours. The ability to conduct experiments in real indoor environments obviously is also highly valuable. Validation of the newly developed protocol for experimentation in residential rooms allows for study of some variables that cannot easily be approximated in the chamber setting, e.g. the contribution to sorption of the many small items that are not included in controlled chamber experiments, and variations by room type (e.g. bathrooms vs. kitchens vs. bedrooms).

The parameter values developed in this study may be used to model sorption in residential and other occupied environments with similar surfaces (e.g. commercial buildings, office spaces, etc.). Sorption rates and partitioning may be estimated for additional compounds using the relationship between the sorptive properties and vapor pressures of the study compounds.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.atmosenv.2006.07.056.

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